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Production of Complex Ceramic Films using domestic Inkjet Printers

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Abstract. The production cost is one of the issues for some complex ceramics applications, like the superconducting oxides. The main properties of such materials, i.e., zero electrical resistivity and perfect diamagnetism, make them attractive for several applications, including energy storage. Thus, in this work, we focus on the production and structural characterization of $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10}$ (Bi-2223) superconducting films using a domestic inkjet printer. The precursor solution was prepared following the Pechini's method, and it was used, such as the ink. Then, an E-shape film was printed over a SiO_2 substrate. The results show that the sample produced with 12 depositions presented a superconducting transition at 81 K and a critical current density of 9.68 A/cm^2 at 78 K.
Keywords: superconducting films, direct printing, BSCCO system.

1. Introduction

The non-dissipative transport of currents at temperatures of 77 K makes the high-temperature superconductors (HTS) fascinating materials for the development of novel devices and their applications at high magnetic fields and in storage energy [1-13]. A large number of those applications consists in the production of HTS films which has been reported to be prepared by sputtering [14,15], PLD [16], and MOCVD [17] techniques. However, to reduce costs and be scalable in the production of films and long-length coated conductors, chemical solution deposition (CSD) techniques have been applied by some researchers [18-26].

Among the HTSs, one of the most studied materials is the Bi-Sr-Ca-Cu-O (BSCCO) system. It was discovered by H. MAEDA and coworkers [27], and the samples present critical temperatures ranging from 7 K to 110 K depending on the superconducting phase

[28-30].

Direct printing is a versatile and fast method among the low-cost CSD techniques to produce films, tapes, and coated conductors of ceramic materials [31-33]. In such a technique, the precursor solution is the ink used to be deposited over the substrate. In some studies, trifluoroacetate (TFA)-based solutions have been used as the chemical ink [34-36].

Then, focusing on alternative CSD methods to produce ceramic materials, in the present study, we describe the fabrication of BSCCO ceramic films using a direct print of the precursor solution using a domestic inkjet printer. General layouts have been printed to show the versatility of the process. We present the characterizations were carried out by x-ray diffraction (XRD) and scanning electron microscopy (SEM) in an E-shape sample. The superconducting properties were analyzed by R(T) and I(V) curves.

2. Materials and Methods

The samples' synthesis was divided into a few steps to demonstrate the versatility of the employed technique as follows.

The layout of the superconducting track

The desired layout can be created on a computer using vector images. As a substrate, we used SiO_2 wafers cut in squares of 2 cm, which was aligned correctly in the printer to replace the paper. Then, the E-shape track was directly printed, as shown in Figure 1.

Production of the precursor solution

The precursor solution was prepared following Pechini's method [37-39], and the stoichiometric calculations were based on the $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10}$ composite, where the presence of Pb is to allow the stabilization of the Bi-2223 phase [40]. The used reagents were: Bi_2CO_5, $SrCO_3$, $CaCO_3$, $CuCO_3.Cu(OH)_2$, $2PbCO_3.Pb(OH)_2$, $C_6H_8O_7$, $C_2H_8N_2$, and $C_2H_6O_2$ (all from Vetec).

The metallic carbonates (MC) were dissolved in an aqueous solution of citric acid ($C_6H_8O_7$) (CA) in a ratio of 3:1 (MC:CA), and after that, the ethylene glycol ($C_2H_6O_2$) was added. It was used 40 w% of citric acid and 60 w% of ethylene glycol. The solution was heated at 80ºC-90ºC to dry until it reached the desired viscosity of about 10.60 cP at 28 ºC, a typical value for the printer's inks.

Heat treatment

The number of depositions was varied to obtain the complete recovery of the substrate. After each deposition, the samples were calcined at 500 °C for 5 minutes using a rate of 2°C/min to eliminate the organic compounds. The sinterization was performed at 835 °C for 10 minutes, using the same heating rate in the air. The final samples are listed in Table 1, which contains the labels and number of depositions.

Table 1 – Samples' label and the number of depositions.

Samples' label	# depositions
T-01	1
T-03	3
T-06	6
T-09	9
T-12	12

Characterizations

The analysis of the structural properties of the samples was made by optical microscopy imaging first to observe the homogeneity of the printed solution and the substrate recovery. X-ray diffraction (XRD) was used to identify the crystalline phases formed after the sinterization process, and scanning electron microscopy (SEM) was used to verify the microstructure of the samples. The electrical properties were measured using the DC four-probe method. From R(T) curves, the critical temperature Tc was obtained, and from the V(I) curves, the value of the critical current of the material.

3. Results and discussions

Figure 1 shows an image taken with an optical microscope of the as printed T-03 sample with a total size of (11x11) mm^2 to exemplify the printed E-type layout. From the zoom of such an image, it is worth noting that the layout was precisely printed with 1mm tracks spaced by 1mm. However, the substrate was not wholly covered, indicating that more depositions should be done.

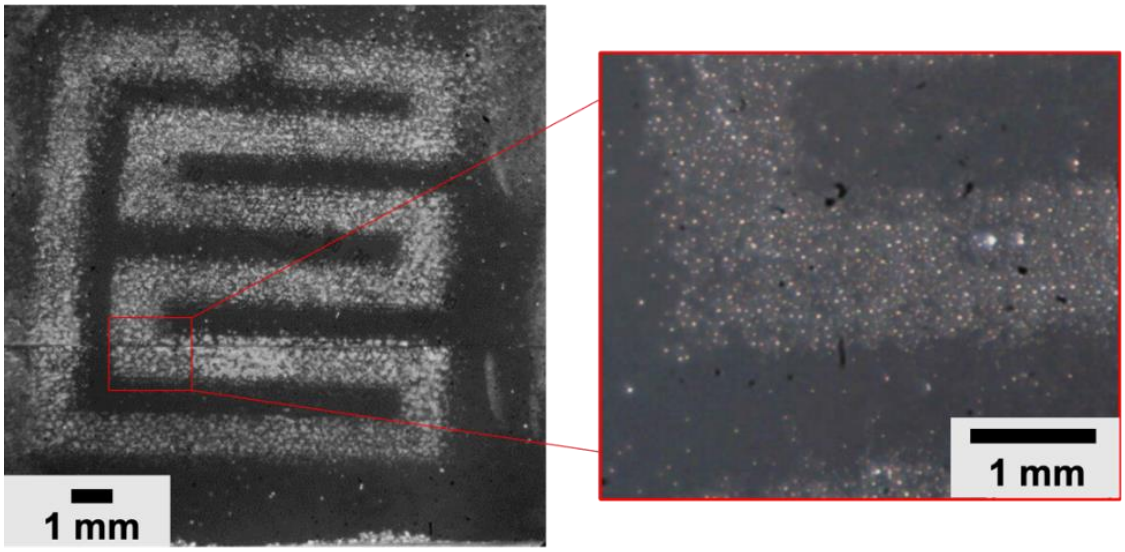

Figure 1 - Optical microphotography of the printed track T-03. The zoom shows the distribution of the droplets along the track.

To follow the substrate's recovery and several depositions, in Figure 2 are shown images taken from an optical microscope after the heat treatment of the samples for the amplification of 3.2x and 12.5x. It can be noted that a considerable portion of the substrate is recovered for samples **T-09 and T-12**.

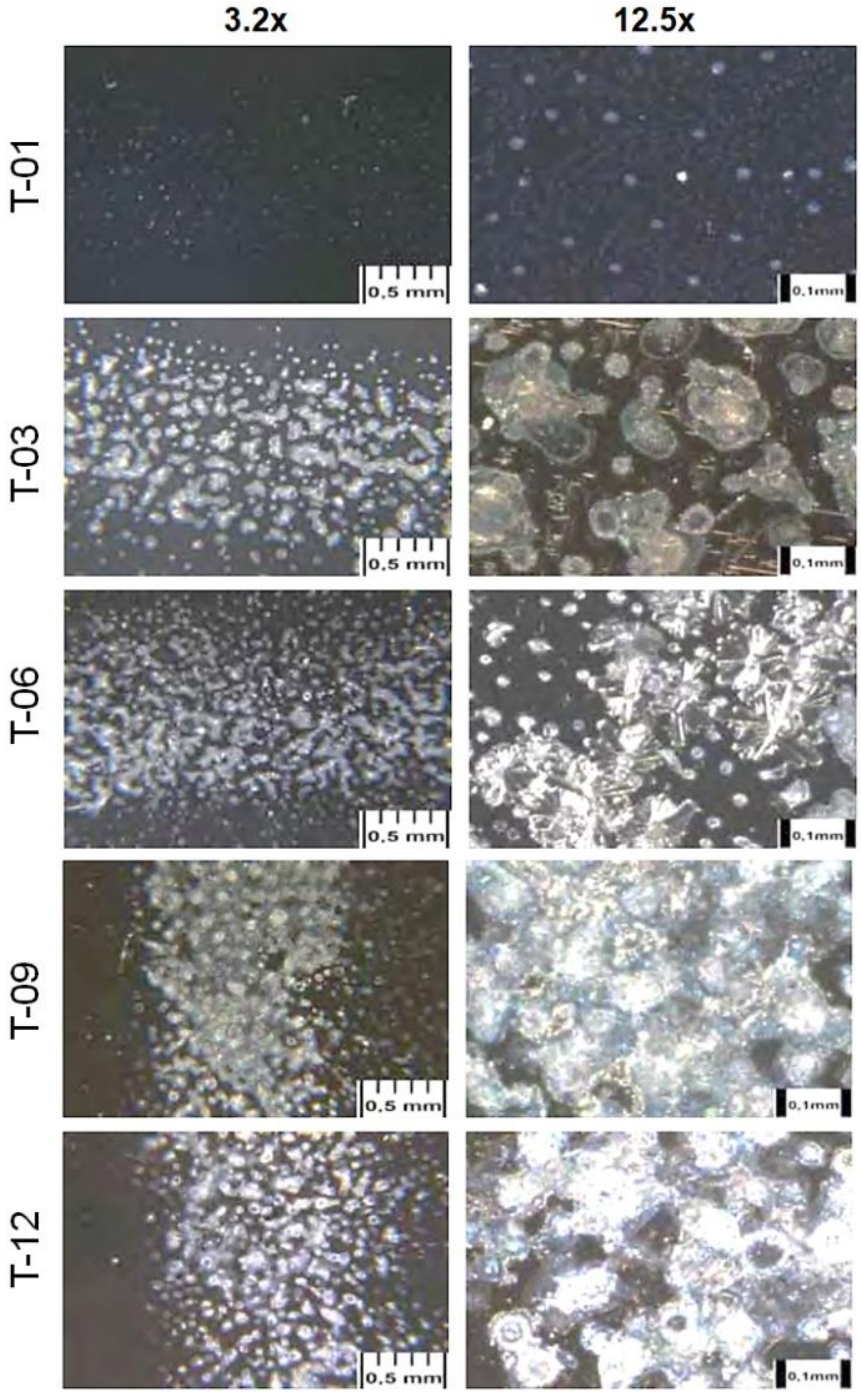

Figure 2 - Optical images of the heat-treated samples after some depositions. The substrate was totally/partially recovered at samples T-09 and T-12.

Focusing on T-12 samples, in Figure 3(a) are shown SEM images with total substrate recovery. It is noticed that there are regions where the grains grew out of the substrate plane, as evidenced in Figure 3 (b). Such behavior can be associated with the number of depositions and the substrate whose crystallographic plane does not match the planes of BSCCO.

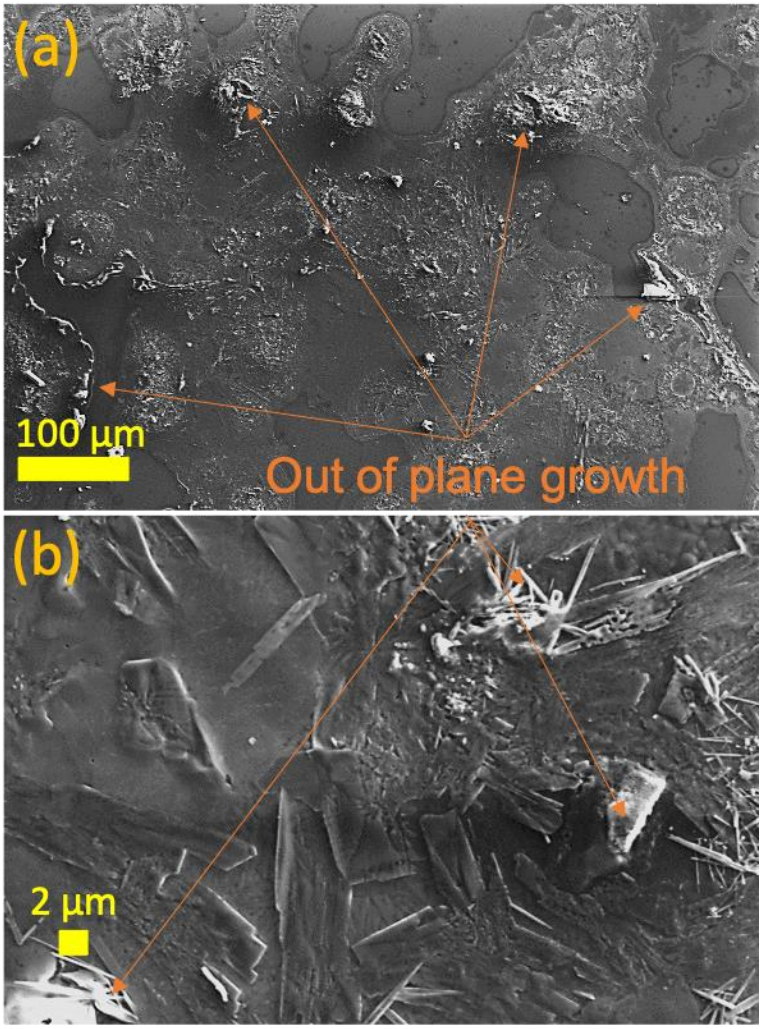

Figure 3 - SEM Images of the samples (a) T-09 and (b) T-12. It is shown the total recovery of the substrate with some structures grown out of the plane.

According to diffractograms, shown in Figure 4 (a) and (b) for T-09 and T-12, respectively, one can note that phases Bi-2223 and Bi-2212 were formed. However, Bi-2212 is more evident [29]. As the film presents a significant area compared with its thickness, part of the Pb presented in the solution evaporates during the heat treatment, facilitating the growth of the Bi-2212 phase over the Bi-2223. The characteristic SiO_2 peak is seen near 33º.

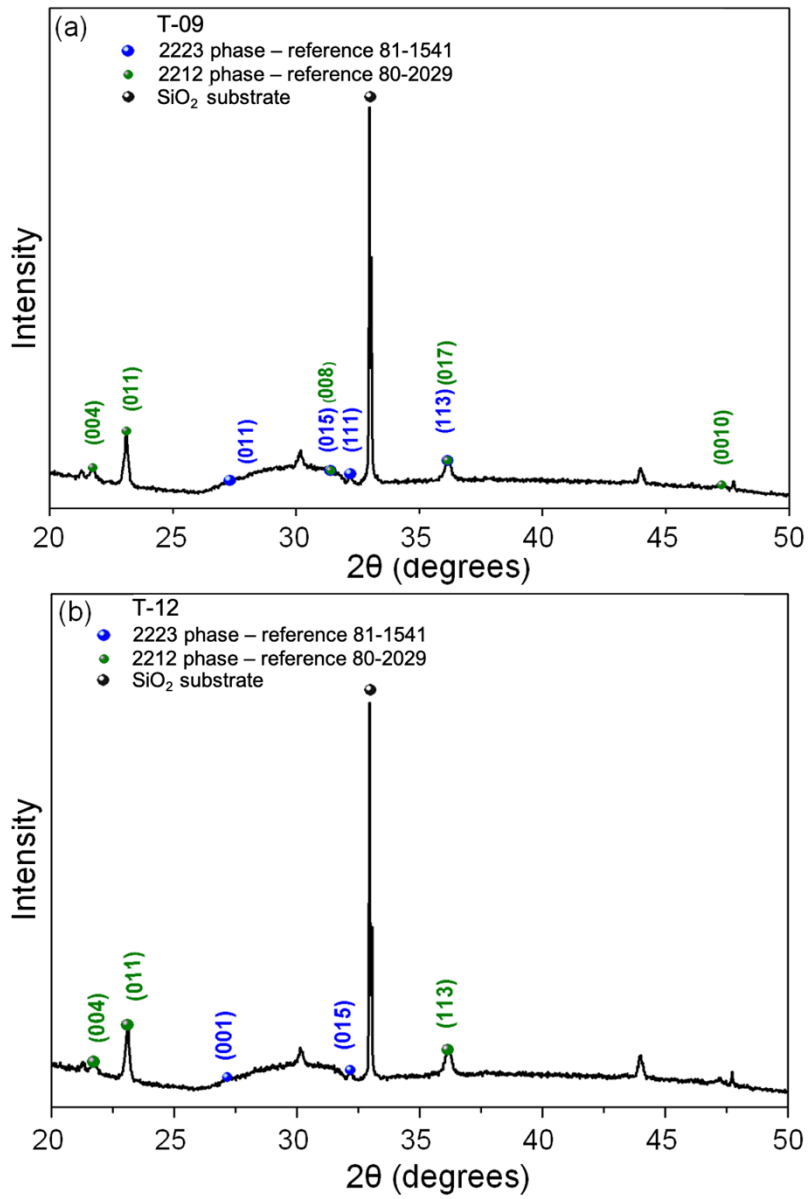

Figure 4 - X-ray diffraction of the samples (a) T-09 and (b) T-12. It can be noted the SiO_2 substrate peak near 33º.

The substrate was not completely covered in samples T-01 and T-03, and no superconducting transition was verified. The T_c of the sample T-12 was (81±1) K. Figure 4 shows the T-12 R(T) curve. Temperatures lower than 77 K cannot be reached with our equipment. The value of T_c indicates that the Bi-2212 is the predominant phase, agreeing with the XRD curves.

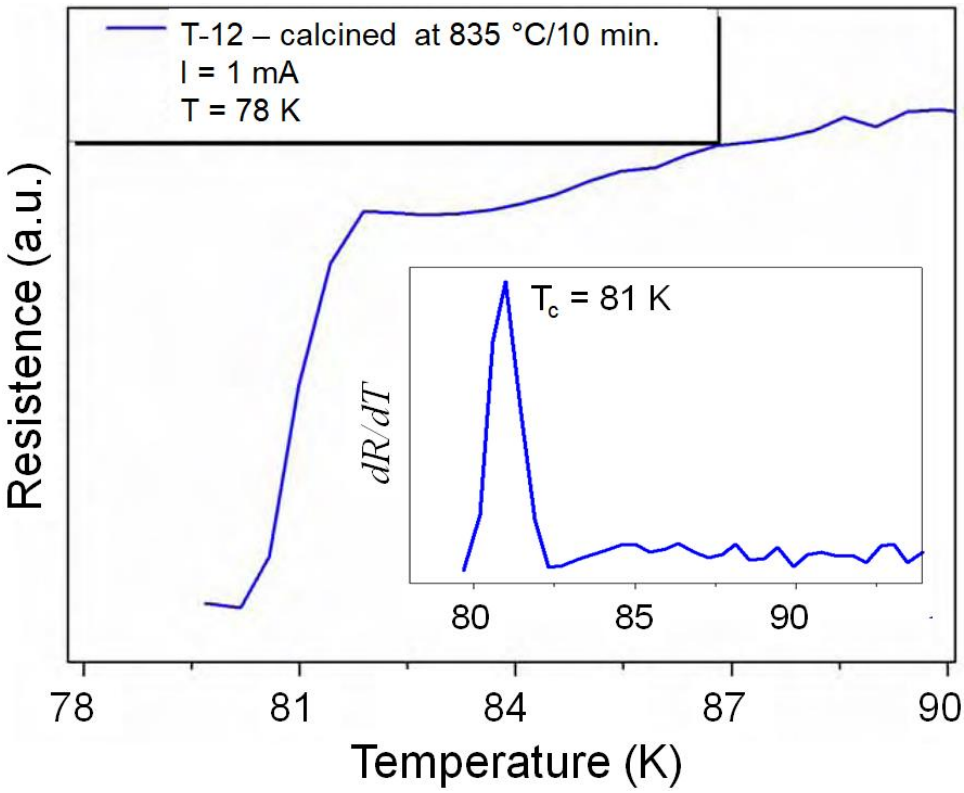

Figure 5 - Resistance as a function of the temperature of the sample T-12.

The critical current, I_c, of T-12 was obtained from V(I) measurement at 78K, shown in Figure 6. Using the 1μV criterion, one obtained I_c= 3.001 x 10^{-4} A. By SEM images (not shown), the thickness of the sample was measured as 3.1 mm, and a width of 1.1 mm. Then, the estimated critical current density was J_c = 9.68 A/cm^2.

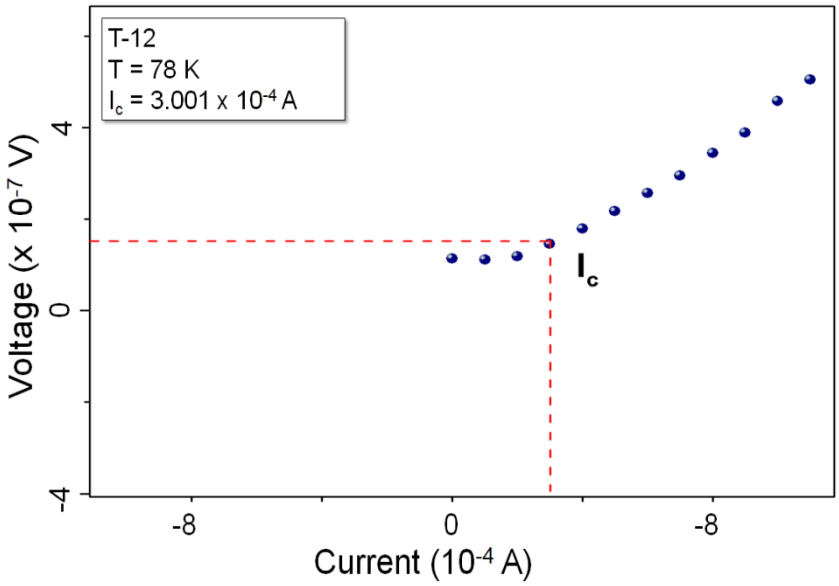

Figure 6 - Characteristic voltage as a function of the electric current of the sample T-12.

4. Conclusions

With a domestic printer and using the precursor solution in the place of the ink, we produced a BSCCO film with a specific layout. Images from an optical microscope confirmed the excellent resolution of the printed layout. SEM images show the growth of plate-like grains, as expected for BSCCO. XRD and electrical characterizations show the excellent quality of the samples. The easy operation and the versatility of the direct print method presented in this work should be applied to various materials and other areas.

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